PB88-222542

Regional Chemical Characteristics of Lakes in North America. Part 2 Eastern United States

(U.S.) Environmental Monitoring Systems Lab.
Research Triangle Park, NC

1986

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TECHNICAL REPORT DATA
(Please read Instructions on the reverse before completing)

1. REPORT NO.	2.	3. PB88-222542
EPA/600/J-86/465		

4. TITLE AND SUBTITLE	5. REPORT DATE
Regional Chemical Characteristics of Lakes in North America; Part II: Eastern United States (Journal Version)	1986
	6. PERFORMING ORGANIZATION CODE
7. AUTHOR(S)	8. PERFORMING ORGANIZATION REPORT NO.
R.A.Linthurst, D.H.Landers, J. Eilers, P.E.Kellar, D.F. Brakke, D.S.Jeffries	
9. PERFORMING ORGANIZATION NAME AND ADDRESS	10. PROGRAM ELEMENT NO.
USEPA;Northrop Services; State U of NY; Radian Corp; Western WU; OSU Corvallis; Envir. Sciences Div.-Oak Ridge,TN; NWRI, Ontario,Canada	11. CONTRACT/GRANT NO.
12. SPONSORING AGENCY NAME AND ADDRESS	13. TYPE OF REPORT AND PERIOD COVERED
US Environmental Protection Agency Environmental Research Laboratory 200 SW 35th Street Corvallis, OR 97333	14. SPONSORING AGENCY CODE EPA/600/2

15. SUPPLEMENTARY NOTES
1986. Water, Air and Soil Pollution 31:577-591.

16. ABSTRACT

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17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group

REPRODUCED BY
U.S. DEPARTMENT OF COMMERCE
National Technical Information Service
SPRINGFIELD, VA. 22161

18. DISTRIBUTION STATEMENT	19. SECURITY CLASS (This Report)	21. NO. OF PAGES
Release to Public	Unclassified	16
	20. SECURITY CLASS (This page)	22. PRICE
	Unclassified	

EPA Form 2220-1 (9-73)

EPA/600/J-86/465
JOURNAL ARTICLE

REGIONAL CHEMICAL CHARACTERISTICS OF LAKES IN NORTH AMERICA

PART II: EASTERN UNITED STATES

by

R. A. Linthurst[1], D. H. Landers[2], J. M. Eilers[3], P. E. Kellar[4],
D. F. Brakke[5], W. S. Overton[6], R. Crowe[7], E. P. Meier[7]
P. Kanciruk[8] and D. S. Jeffries[9]

[1]U. S. EPA, Environmental Monitoring Systems Laboratory, MD-39, Research Triangle Park, NC 27111, USA

[2]State University of New York, King Hall, 300 Washington Blvd., Oswego, NY 13126 USA Present Address: U.S. EPA, Environmental Research Laboratory, 200 S. W. 35th Street, Corvallis, OR 97333, USA

[3]Northrop Services, Inc., 200 S. W. 35th Street, Corvallis, OR 97333, USA

[4]Radian Corporation, P.O. Box 13000, Research Triangle Park, NC 27709, USA

[5]Western Washington University, Institute for Watershed Studies, Bellingham, WA 98225, USA

[6]Oregon State University, Department of Statistics, Kidder Hall, Corvallis, OR 97333, USA

[7]U.S. EPA, Environmental Monitoring Systems Laboratory, Las Vegas, NV 89114, USA

[8]Environmental Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN 37830, USA

[9]National Water Research Institute, Box 5050, Burlington, Ontario L7R 4A6, Canada

Water, Air, and Soil Pollution **31** (1986) 577–591.
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ABSTRACT. A survey of 1592 lakes (>4 ha and ≤2,000 ha) was conducted in Fall, 1984. The study area was three regions of the eastern United States (Northeast, Upper Midwest, and Southeast) expected to contain the most lakes with acid neutralizing capacity (ANC) ≤400 μeq/L. The design of the survey provides statistically reliable estimates of the number, location and chemical characteristics of lakes in the study area. Frequency distribution statistics are presented for pH, ANC, sulfate and organic anion. The highest percentages and numbers of acidic lakes occurred in Florida, the Adirondacks, and the Upper Peninsula of Michigan. The highest percentages and numbers of lakes with high sulfate concentrations occurred in the Poconos/Catskills, Southern New England and Florida. The highest percentages and numbers of lakes with high organic anion concentrations occurred in Northeastern Minnesota, the Upper Great Lakes Area and Florida. The percentage of acidic lakes by sulfate concentration class revealed no discernable pattern across subregions. In contrast, the acidic lakes occur with the highest frequency in the lowest organic anion concentration class. The complex distribution of low ANC lakes cannot be explained simply by sulfate concentrations or organic acids.

1. INTRODUCTION

Despite several attempts to quantify change related to acidic deposition, the geographical extent of lakes and streams potentially at risk, or acidic, and the degree or severity to which North American surface waters as a whole are threatened by acidic deposition, have remained unresolved. Considerable statistical uncertainty in extrapolating the results of specific studies to a regional or national scale has continued to exist largely due to: the unknown degree to which the individual study sites represented the larger regional population; bias in lake selection; the absence of measurements of key chemical variables which are considered critical in assessing effects; the difficulty in inter-study comparisons of data collected by different investigators; non-standard methodological techniques; and the variable documentation of quality assurance protocols.

The objective of this paper is to explore the chemical relationships among lakes receiving sulfate loading from atmospheric deposition in the eastern United States, using a uniformly generated data base that was subjected to rigorous quality assurance protocols (Linthurst *et al.*, 1986; Overton *et al.*,1986; Kanciruk *et al.*, 1986). Specifically, the questions posed here are: 1) do pH and concentrations of ANC, sulfate and organic anion in lakes show patterns that can be distinguished regionally? 2) are sulfate concentrations in lakes inversely related to ANC concentrations? 3) are increased organic anion concentrations directly related to ANC concentrations? These are addressed with the assumption that atmospheric deposition of strong acids, principally sulfuric acid, can produce acidic lakes. Of interest in our analysis is to determine what patterns are regional in nature, what relationships are not valid on a regional scale, and to define the specific sub populations of lakes where acidic deposition effects are observed.

2. METHODS

Detailed methods are given in Linthurst et al. (1986). Methods bearing on interpretations are summarized below.

2.1. Population Sampled

Lakes were selected using a stratified design with equal allocation of sample lakes among strata (Linthurst et al., 1986). Three stratification factors were used: region, subregion and alkalinity map class. Lakes were selected in each stratum by systematic sampling of an ordered list following a random start.

Three geographical regions in the eastern United States (Figure 1) were chosen for study during the first phase of the Eastern Lake Survey: the Northeast, Upper Midwest, and Southeast. The regions were chosen on the basis of alkalinity maps generated from historical data by Omernik and Powers (1983). The lakes within these regions were expected to be characterized predominantly by ANC ≤ 400 μeq/L. Subregions (Figure 1) were delineated on the basis of similarities in surrounding physical geography, land use characteristics, and available water quality information.

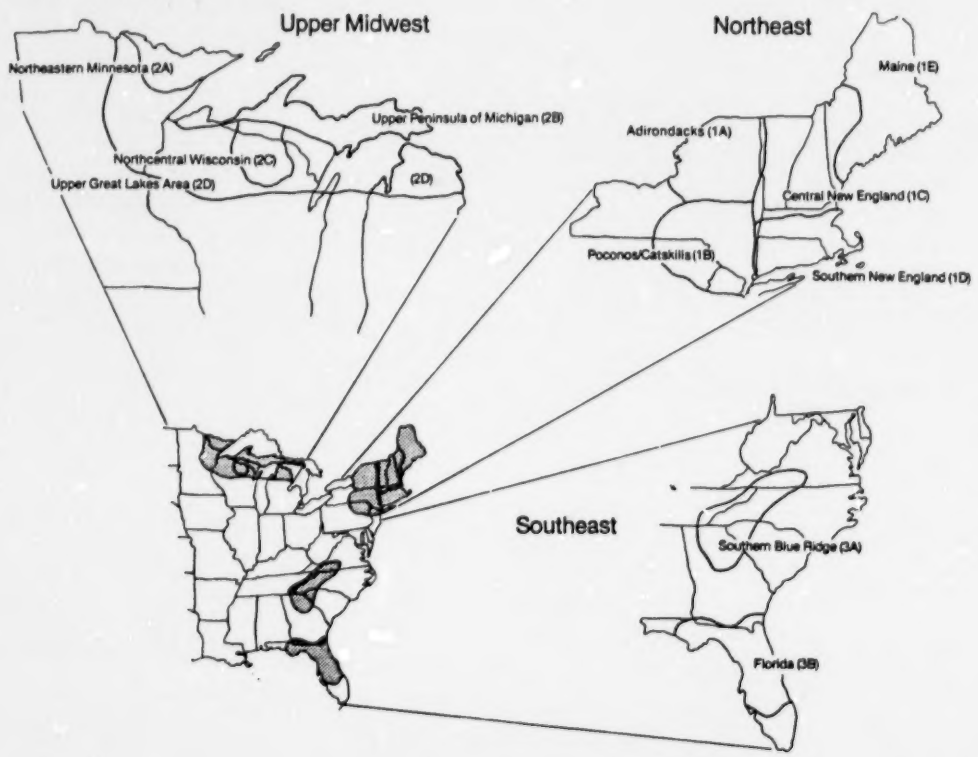

Figure 1. Regions and subregions surveyed in Phase I of the Eastern Lake Survey.

Each subregion was further divided into three smaller areas using the alkalinity maps of Omernik et al. (1985), Omernik and Kinney (1985) and Omernik and Griffith (1985). Areas within each subregion were delineated such that three classes of lakes with expected alkalinities of <100, 100 to 200 and > 200 μeq/L were distinguished. The lakes to be sampled within each alkalinity class were drawn from a preliminary map population list so that each had a known probability of being chosen for sampling. An approximately equal number of lakes (about 50) was chosen from each class.

The target population within the three regions was based on 1:250,000 scale U.S. Geological Survey (USGS) maps. Because such maps in the eastern U.S. usually do not include lakes with surface area less than approximately 4 ha, these smaller lakes are not represented in the target population. The target population was refined before sampling using large-scale (e.g., 1:24,000) USGS maps, during sampling using observations by the sampling teams and following sampling based on evaluation of the data. The final target population excluded lakes which were <4 ha, non-lakes (dry or flowing water), wetlands, high conductance (>1500 μS/cm), impacted by anthropogenic sources, inaccessible or frozen. All population estimates reported in this paper refer to the target population.

2.2. Sampling Methods

Sampling was conducted using helicopters equipped with floats (Morris et al., 1986), during fall turnover when most of the lakes were unstratified. Sampling periods were October 8 to November 8, 1984 in the Northeast, October 17 to November 13, 1984 in the Upper Midwest, and November 17 to December 14, 1984 in the Southeast. In most lakes discrete samples from 1.5 m were taken from sites generally located in the deepest part of the lake. In a few lakes that were too shallow to permit obtaining a clean sample from 1.5 m, samples were collected at 0.5 m. Lakes greater than 2000 ha were sampled but have been excluded from discussion in this paper.

2.3. Analytical Methodologies

Preliminary sample processing, including filtration and preservation, was conducted at base sites in specially equipped mobile field laboratories usually within 12 h of collection. Samples were then shipped by overnight air to one of four analytical laboratories where analyses were completed within pre-established holding times specific to each chemical variable. A total of 25 analytical measurements as made in situ, in the field laboratory and in the analytical laboratories (Hillman et al. 1986). Results for pH, ANC, sulfate and organic anion are presented here.

An extensive quality assurance program was implemented during the ELS -I to ensure that all sampling and analytical protocols were standardized (Best et al., 1986). The data were subjected to two primary quality assurance checks--verification and validation--conducted by the EPA Environmental Monitoring Systems Laboratory at Las Vegas, NV

(Drouse et al., 1986) and the EPA Environmental Research Laboratory at Corvallis, Oregon (Eilers et al., 1986).

pH was determined electrometrically in the field laboratory on samples collected in gas-tight syringes using a special chamber to avoid exchange of dissolved carbon dioxide with air (Hillman et al., 1986). ANC was determined in an analytical laboratory using a modification of the Gran technique (Hillman, ibid). Sulfate was determined using ion chromatography. Sulfate data were not corrected for sea salt because of the influence of road salt in some areas.

Dissolved organic carbon (DOC) was determined by flame ionization detector spectrophotometry. Organic anion concentrations were calculated using DOC and H^+ concentrations according to the model equations described in Oliver et al. (1983). This model was developed using data from colored waters and assumes that most of the organic carbon is humic material. The organic anion values for this paper were calculated using DOC data from both colored and clear lakes and, consequently, may be overestimates.

3. RESULTS

3.1. Regional Characterization - pH and ANC

Within the eastern U.S., median pH values ranged from a low in Subregion 3B (Florida) of 6.6 to a high of 7.4 in the Upper Great Lakes Area (2D) in the Upper Midwest (Table I). The lowest Q_1 values were observed in the Adirondacks subregion (1A) and Florida. The highest percentages of lakes with pH $\leq$ 5.0 occurred in Adirondacks subregion, the Upper Peninsula of Michigan (2B) and Florida (3B). Relatively few lakes in the target population in the Eastern U.S. were characterized by having low pH ($\leq$ 5.0).

In the Northeast, median pH values were fairly similar among subregions. The median values ranged from 6.7 in Subregion 1A to 7.0 in Subregion 1B (Poconos/Catskills), and the Q_4 values ranged from 7.1 in Subregions 1A and 1D to 7.4 in 1B. In Maine (1E) 66.8% of the lakes had ANC $\leq$ 200 µeq/L but only 0.5% had negative ANC and pH $\leq$ 5.0.

In the Upper Midwest, the lowest median pH (6.7) and ANC (93.9 µeq/L) occurred in Northcentral Wisconsin (2C). Over 98% of the lakes in Northeastern Minnesota (2A) which contains the Boundary Waters Canoe Area, had pH values > 6.0. The highest percentage (nine) of low pH lakes ($\leq$ 5.0) occurred in the Upper Peninsula of Michigan (2B) as did the highest percentage of lakes with ANC $\leq$ 0 µeq/L (9.8%). As in Subregion 2A, nearly all the lakes in Subregion 2D had pH values $\leq$ 6.0 and less than one-third had ANC $\leq$ 200 µeq/L.

Florida (3B) had the lowest median pH and ANC of any subregion in the U.S. All of the lakes in the Southern Blue Ridge (3A) had pH values $\geq$ 5.5; no lakes with ANC $\leq$ 0 µeq/L were sampled. In contrast, 12.4% of the Florida lakes had pH values $\leq$ 5.0 and 22.0% had negative ANC.

Table I

Median, Q_1[a] and Q_4[a] values for pH (Table I) and ANC (Table II) in the eastern United States. Percentages of lakes sampled which fall into three pH (Table I) and ANC (Table II) classes are given. All values are target[b] population estimates. The 95% upper confidence limit for the percentage is given in parentheses.

		pH Values			Population Estimates(%)[c]		
	$\hat{N}$	Q_1	Med.	Q_4	$\leq$5.0	$\leq$5.5	$\leq$6.0
1A	1290	5.5	6.7	7.1	10.0(14.0)	19.9(25.1)	26.6(32.4)
1B	1479	6.6	7.0	7.4	0.8(1.3)	5.7(9.9)	7.8(12.1)
1C	1483	6.3	6.8	7.3	1.7(3.2)	7.8(11.2)	12.9(17.2)
1D	1318	6.2	6.8	7.1	5.0(8.7)	10.0(14.7)	14.6(19.8)
1E	1526	6.6	6.9	7.3	0.5(1.3)	1.6(3.0)	4.8(7.2)
1	7096	6.3	6.9	7.3	3.4(4.5)	8.6(10.3)	12.9(14.9)
2A	1457	6.6	6.9	7.3	0.0(-)	0.0(-)	1.4(3.0)
2B	1050	6.1	7.1	7.8	9.4(14.6)	13.6(19.4)	17.7(24.1)
2C	1480	5.8	6.7	7.5	2.1(3.7)	11.1(14.7)	27.7(33.0)
2D	4515	6.6	7.4	8.1	0.0(-)	0.1(0.2)	4.5(8.8)
2	8501	6.4	7.1	7.9	1.5(2.2)	3.6(4.5)	9.6(10.0)
3A	258	6.7	7.0	7.3	0.0(-)	0.0(-)	0.4(0.5)
3B	2098	5.4	6.6	7.4	12.4(18.4)	20.6(27.3)	32.7(41.8)

Table II

		ANC (μeq L^{-1})			ANC (μeq L^{-1}) Population Estimates (%)[c]		
	$\hat{N}$	Q_1	Med.	Q_4	$\leq$0	$\leq$40	$\leq$200
1A	1290	14.0	111.8	250.8	10.7(14.8)	31.3(37.4)	70.5(77.2)
1B	1479	102.2	297.4	530.5	5.3(9.5)	12.2(18.1)	38.7(48.6)
1C	1483	52.0	119.9	387.7	2.4(4.3)	16.4(21.1)	67.6(74.2)
1D	1318	39.4	161.8	402.6	5.0(8.1)	19.6(25.5)	57.3(66.9)
1E	1526	72.1	148.4	298.4	0.5(1.3)	7.6(10.7)	66.8(73.4)
1	7096				4.6(6.0)	17.0(19.3)	60.0(63.6)
2A	1457	98.2	184.9	403.4	0.0(-)	1.9(4.1)	57.0(64.5)
2B	1050	55.5	283.6	1284.4	9.8(15.0)	18.3(24.6)	41.7(51.0)
2C	1480	16.8	93.9	600.7	3.1(5.0)	37.0(43.1)	56.7(63.3)
2D	4515	104.0	801.7	2021.9	0.0(-)	3.9(7.1)	31.3(40.8)
2	8501				1.7(2.4)	11.1(13.3)	41.4(46.9)
3A	258	149.1	250.2	421.4	0.0(-)	0.8(1.1)	34.3(41.9)
3B	2098	-7.5	83.5	713.0	22.0(29.3)	32.9(37.5)	55.1(67.3)

[a]It is estimated that 20% of the lakes have values less than or equal to Q_1 and 80% are less than Q_4.

[b]Target population size ($\leq$ 2000 ha) was estimated by weighting sample n for the probability of the lake being sampled.

[c](-) = undefined

3.2 Regional Characterization - Sulfate Concentrations in Lakes

In general, median sulfate concentrations in lakes were higher in the Northeastern U.S. than in the Upper Midwest (Table III). In the Southeast, the median concentration in Florida was within the range observed in the Northeast while the Q_4 concentration (272 μeq/L) was the highest observed in all three subregions. The lowest median sulfate concentration (32 μeq/L) occurred in the Southern Blue Ridge (3A).

Within the Northeast, the subregion containing the Poconos/Catskills Subregion (1B) had the highest median sulfate concentration while Maine (1E) had the lowest. Only 1% of the lakes in Maine had sulfate concentrations >150 μeq/L while 12.8% of the lakes in Subregion 1A had sulfate in excess of 150 μeq/L. The highest percentage of lakes with sulfate concentrations >150 μeq/L occurred in Subregion 1B, followed by Southern New England (1D).

The highest sulfate concentration in the Upper Midwest occurred in the Upper Peninsula of Michigan. The lowest Q_1 and median occurred in Subregion 2D (the Upper Great Lakes Area). In Subregions 2A and 2C nearly all the lakes were characterized by sulfate concentrations >150 μeq/L. Subregion 2D had the highest proportion of lakes with sulfate concentrations >150 μeq/L followed by Subregion 2B.

The Southern Blue Ridge had the lowest median, Q_1 and Q_4 sulfate concentrations and the lowest percentage of lakes with concentrations >50 μeq/L of any U.S. subregion. In contrast, the median concentration in Florida was nearly three times that in Subregion 3A and within the range observed in the Northeast. The Q_4 concentration in Florida was the highest observed in the eastern U.S. Sixty-eight percent of the lakes in Subregion 3B had concentrations >50 μeq/L, while 40.3% of the lakes had concentrations >150 μeq/L.

3.3 Regional Characterization - Organic Anion Concentrations

Generally, the highest organic anion concentrations were observed in the Upper Midwest and Florida (Table IV). The greatest proportions of lakes with organic anion >80 μeq/L occurred in Subregions 2A, 2D and 3B where 49% or more of the lakes fell into this category. The lowest percentage of lakes with high organic anion concentrations was found in the Southern Blue Ridge (3A), where only 2.4% of the lakes had organic anion >80 μeq/L.

Maine had the highest median and Q_4 organic anion concentrations in the Northeast. Five percent or fewer of the lakes in the subregions containing the Adirondacks and the Poconos/Catskills had organic anion >80 μeq/L.

Northeastern Minnesota (2A) had the highest median organic anion concentration in the Upper Midwest. The lowest median concentration occurred in Subregion 2C, which also had the lowest percentage of lakes with organic anion concentrations >80 μeq/L.

In the Southern Blue Ridge (Subregion 3A) the median organic anion concentration was 19.7 μeq/L compared to that in Florida which was 78.2 μeq/L. Only 34.4% of the lakes in 3A had organic anion concentrations >30 μeq/L while in Subregion 3B, 81.6% of the lakes occurred in

Table III

Median, Q_1[a] and Q_4[a] values for lake sulfate (Table III) and organic anion (Table IV) in the eastern United States. Percentages of lakes sampled which fall into three sulfate classes are given. All values are target[b] population estimates. The 95% upper confidence limit for the percentage is given in parentheses.

		Sulfate Concentrations ($\mu eq\ L^{-1}$)			Sulfate ($\mu eq\ L^{-1}$) Population Estimates (%)[c]		
	$\hat{N}$	Q_1	Med.	Q_4	>50	>100	>150
1A	1290	104.0	118.7	141.9	99.4 (100)	84.1(91.0)	12.8(16.8)
1B	1479	118.1	159.3	221.0	99.8 (100)	90.6 (100)	56.1(67.9)
1C	1483	77.9	101.2	136.1	99.5 (100)	51.2(58.1)	15.7(20.2)
1D	1318	108.2	141.1	190.3	100 (100)	87.0(98.6)	45.8(55.9)
1E	1526	56.4	74.6	87.8	88.5 (95.8)	9.3(12.8)	1.1(2.3)
1	7096	79.4	115.4	164.9	97.3 (100)	63.0(66.8)	26.0(29.3)
2A	1457	46.6	62.5	83.9	73.7 (82.7)	9.7(14.4)	1.9(4.0)
2B	1050	49.1	77.7	103.5	79.1 (90.8)	26.3(35.5)	6.6(11.8)
2C	1480	41.9	56.9	76.4	69.5 (76.9)	6.4(9.6)	1.0(2.6)
2D	4515	29.0	50.1	85.4	50.1 (62.0)	16.5(24.9)	11.0(18.0)
2	8501	34.5	57.1	87.8	61.1 (67.9)	14.8(19.5)	7.1(10.9)
3A	258	19.5	31.8	59.6	23.1 (31.7)	14.6(21.6)	8.5(13.9)
3B	2098	28.4	93.7	272.4	68.1 (82.0)	49.4(61.5)	40.3(51.8)

Table IV

		Organic Anion Concentrations ($\mu eq\ L^{-1}$)			Organic Anion ($\mu eq\ L^{-1}$) Population Estimates (%)[c]		
	$\hat{N}$	Q_1	Med.	Q_4	>10	>30	>80
1A	1290	26.1	37.3	53.3	96.6 (100)	73.1(80.2)	4.3(6.9)
1B	1479	28.4	37.3	57.5	97.3 (100)	72.6(84.4)	84.6(8.8)
1C	1483	29.9	42.9	65.4	98.3 (100)	80.0(87.1)	10.9(14.9)
1D	1318	22.3	37.9	65.7	96.0 (100)	62.9(73.9)	11.0(16.5)
1E	1526	28.2	51.5	80.8	98.9 (100)	72.0(84.4)	23.2(28.5)
1	7096	27.3	41.2	64.8	97.5 (100)	73.4(77.5)	11.1(13.1)
2A	1457	54.8	89.0	121.4	100.0 (100)	98.8 (100)	58.5(67.3)
2B	1050	32.0	66.6	111.0	99.6 (100)	83.9(95.5)	33.7(43.4)
2C	1480	29.7	45.6	77.5	98.5 (100)	78.7(85.8)	19.7(25.6)
2D	4515	49.8	81.8	128.0	100.0 (100)	96.3 (100)	52.4(64.3)
2	8501	42.6	72.7	120.2	99.7 (100)	92.1(98.5)	52.1(45.4)
3A	258	11.1	19.7	38.0	85.3 (93.2)	34.4(44.8)	2.4(5.1)
3B	2098	35.7	78.2	130.5	91.1 (100)	81.6(97.6)	49.0(62.4)

[a]It is estimated that 20% of the lakes have values less than or equal to Q_1 and 80% are less than Q_4.

[b]Target population size (< 2000 ha) was estimated by weighting sample n for the probability of the lake being sampled.

[c](-) = undefined

this category.

3.4 Regional Characterization - Relationship Between ANC and Sulfate

The distribution of lakes with ANC $\leq$ 200 μeq/L by sulfate class is shown for the Northeast (Figure 2A) and Upper Midwest and Southeast (Figure 2B). Data are arranged within region by increasing median sulfate for all lakes sampled within the subregion. Five sulfate classes, based on the first through fifth quintiles, were derived from the cumulative frequency distribution of sulfate using the entire data sets from Canada (see Jeffries, et al., 1986) and the U.S. The quintile values represent the maximum concentration below which 20, 40, 60, 80, and 100% of the sample (Canada) and estimated target population (U.S.) would be found. Within each sulfate class, lakes characterized by ANC < 0, 0 to 40, 41 to 100, and 101 to 200 μeq/L are shown. In subregions with the highest overall median sulfate, the distribution of lakes with ANC $\leq$ 200 μeq/L is skewed toward the higher sulfate classes.

In the Northeast, all the lakes with ANC $\leq$ 200 μeq/L within the Poconos and Catskills (1B) and Southern New England (1D) were found in the upper three sulfate classes. In Subregion 1E, which had the lowest population median sulfate concentration in the Northeast, most of the lakes with ANC $\leq$ 200 μeq/L were found in the lower three sulfate classes. The largest percentage of lakes with ANC $\leq$ 40 μeq/L were not consistently found in the highest sulfate classes within each subregion. The distributions of lakes among sulfate classes were more variable in Subregions 1A, 1C and 1E than in 1B and 1D.

In the Upper Midwest, lakes with sulfate concentrations in the fifth class (> 163 μeq/L) occurred only in Northcentral Wisconsin (2C). Very few lakes in Subregions 2A, 2B and 2C occurred within the fourth class. Most of the lakes in the Upper Midwest occurred in the lower three sulfate classes. A greater percentage of lakes with ANC $\leq$40 μeq/L was found in sulfate class 2 than in sulfate class 3 in both Subregions 2B and 2C, both of which contained the highest percentages of low ANC lakes in the Upper Midwest.

In the Southeast, lakes in Florida (3B) were distributed across all sulfate classes. The greatest percentage of lakes with negative ANC was found in the lowest sulfate class, but lakes with ANC $\leq$ 40 μeq/L were found among all sulfate classes. The highest percentage of low ANC lakes was found in the lowest and not in the highest sulfate class. In the Southern Blue Ridge, most of the lakes occurred in the lowest sulfate class with the remaining few occurring in sulfate classes 2 and 5. Those lakes in sulfate class 5 had ANC in the 101 to 200 μeq/L range.

3.5 Regional Characterization - Relationship Between Organic Anion and ANC

The distributions of lakes with ANC $\leq$ 200 μeq/L by organic anion class are shown for the Northeast and the Upper Midwest and Southeast (Figure 3B). Data are arranged within region by increasing median organic anion concentration for all lakes sampled within the subregion. Five organic anion categories were defined in the same manner as the sulfate classes.

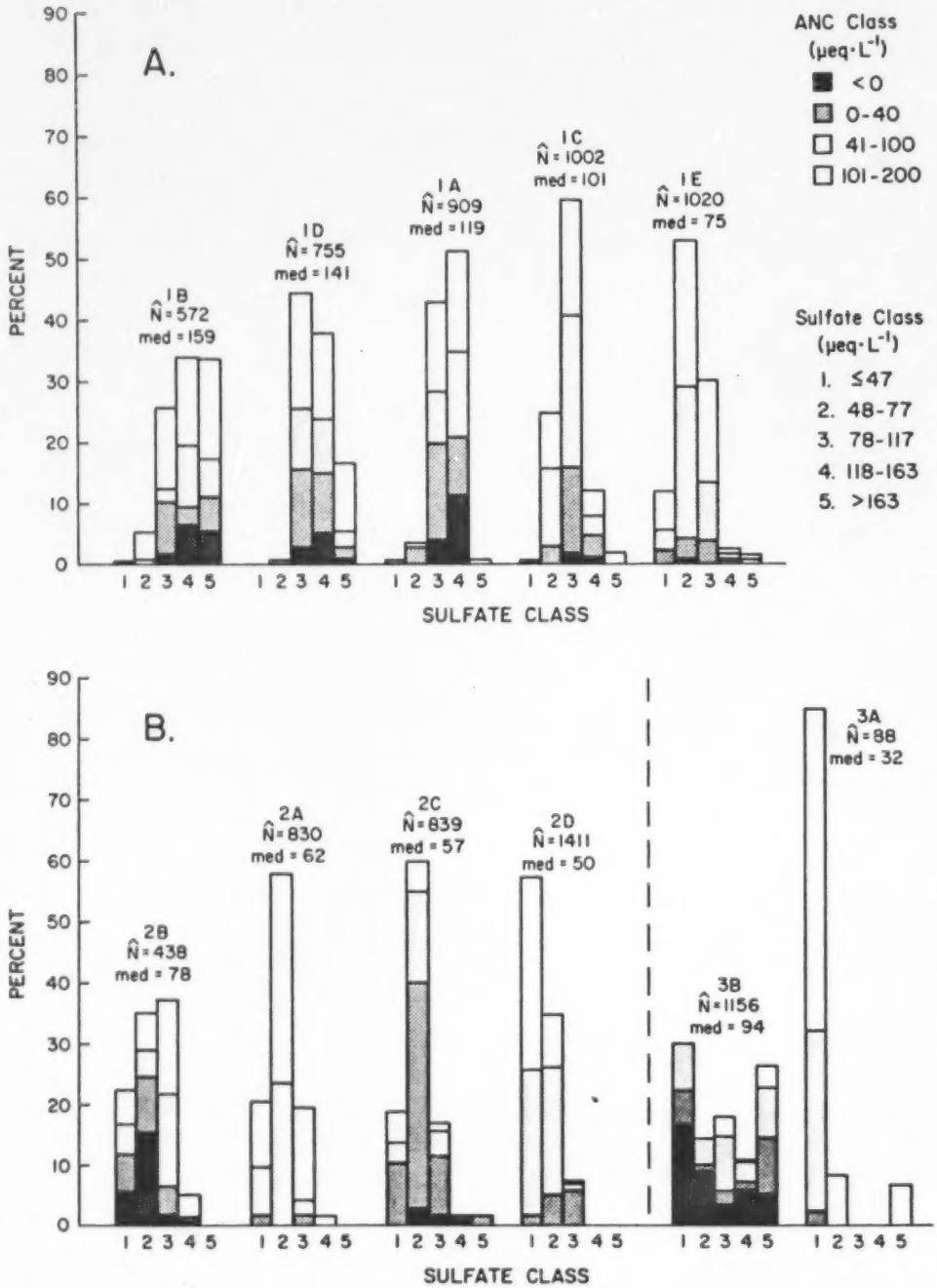

Figure 2. The distribution of lakes with ANC≤200 μeq/L across 5 sulfate classes for the Northeast (A) and the Upper Midwest and Southeast (B). Within each sulfate class, the proportions of lakes characterized by one of four ANC concentration classes are given. Subregions are arranged in order of decreasing overall population median estimates of sulfate concentration.

In the Northeast (Figure 3A) as the overall median organic anion concentration (including lakes with ANC $\leq$ 200 μeq/L) decreased, the distributions generally tended to shift to the lower organic anion classes. The distributions within Maine (1E) and Central New England (1C) were similar with a slightly higher percentage of lakes occurring in the lowest organic anion class in Subregion 1C, which had a lower overall median organic anion concentration. The distributions within Subregions 1B, 1A, and 1D were somewhat different despite the fact that the overall median values were ~37 μeq/L in each of these subregions. In each of these three subregions, proportionally more lakes were found in the lower organic anion classes than in Subregions 1E and 1C.

The majority of lakes with negative ANC were found in the lowest organic anion class. In general, no relationship between increasing organic anion concentration and decreasing ANC concentration was observed; that is, as organic anion increased, the percentages of lakes characterized by ANC $\leq$ 40 μeq/L did not increase. In fact, with increasing organic anion, ANC tended to increase.

In the Upper Midwest and Southeast, as the overall median organic anion decreased, the percentages of lakes with ANC $\leq$ 200 μeq/L generally shifted from the upper to the lower organic anion classes. The distributions in Northcentral Wisconsin (2C) and Florida (3B) were bimodal with relatively few lakes in the middle organic anion class (38-50 μeq/L). In Northeastern Minnesota (2A), which had the highest overall median organic anion concentration, most of the lakes with ANC $\leq$ 200 μeq/L were characterized by >73 μeq/L organic anion. Most of the lakes with ANC $\leq$ 200 μeq/L in the Southern Blue Ridge (3A) were characterized by organic anion concentrations >26 μeq/L. As in the Northeast, lakes with negative ANC were generally characterized by organic anion concentrations in class 1 and class 2. In the Upper Peninsula of Michigan (2B) and in Florida (3B), a small percentage of lakes with negative ANC were found in organic anion classes 4, and 4 and 5, respectively.

4. DISCUSSION

Concentrations of ANC, sulfate, and pH in lakes of the eastern U.S. exhibit high variability within and among subregions. This regional heterogeneity in the eastern U.S. underscores the difficulty in drawing direct inferences between the occurrence of acidic lakes and atmospheric deposition. For example, the Upper Peninsula of Michigan (2B) and the Upper Great Lakes Area (2D) lie along a similar depositional gradient, and yet 2D contains virtually no lakes with pH $\leq$ 5.0 whereas 9.4% of the lakes in 2B have pH $\leq$ 5.0. In this case, terrigenous properties of the watersheds strongly influence lake chemistry. Certain patterns are apparent, however, that suggest a distribution in lake chemistries among subregions that may be related to acidic deposition. For example, the Northeast, Southern New England, the Poconos/Catskills, and the Adirondacks subregion receive high loadings of sulfate deposition, whereas northern New England and most of Maine receive substantially less (U.S./Canada, 1983). The subregional sulfate concentrations correspond with this pattern in sulfate deposition for the Northeast and Upper Midwest.

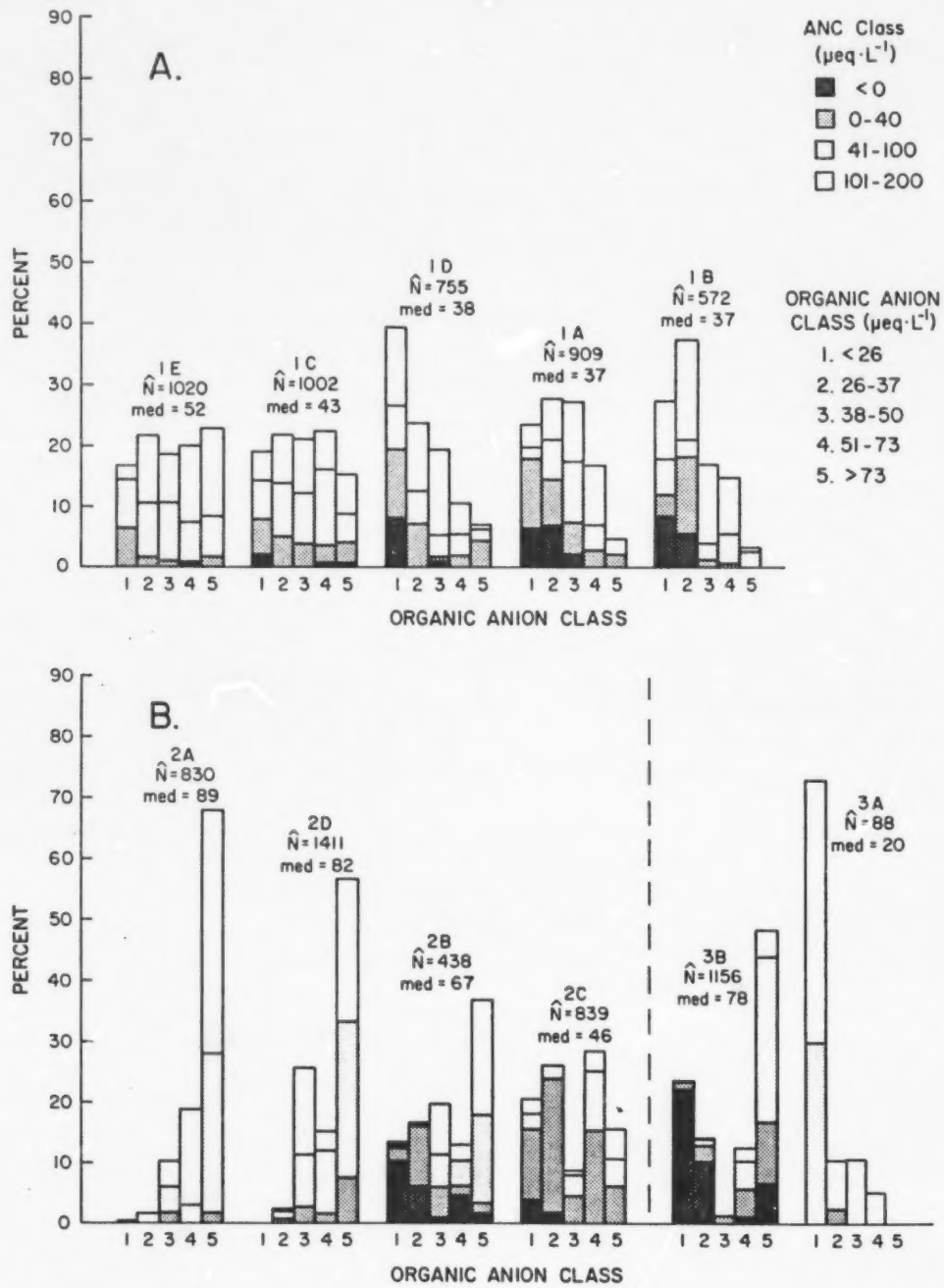

Figure 3. The distributions of lakes with ANC≤200 μeq/L across each of 5 organic anion classes for the Northeast (A) and the Upper Midwest and Southeast (B). Within each organic anion class, the proportions of lakes characterized by one of four ANC classes are given. Subregions are arranged in order of decreasing overall population median estimates of organic anion.

The data in this survey and the companion Canadian compilation (Jeffries et al., 1986) indicate that: (1) in-lake ANC concentrations (or pH) are not related directly to sulfate concentrations; and (2) most acidic lakes are not caused by naturally occurring organic acids. The absence of an obvious relationship between sulfate and ANC within the subregions reflects a variety of non-conservative watershed processes and multiple sulfate sources influencing lake concentrations, as well as differences in sources of ANC. These processes include sulfate adsorption by soils, biological uptake by vegetation, groundwater contributions from sulfide minerals or marine deposits, sea salt contributions, in-lake sulfate reduction, and alkalinity generation. Further stratification of the lakes by relevant ecological criteria may reveal a stronger relationship between ANC and sulfate (e.g., in clearwater lakes within uniform geological terrain). The apparently complex relationship between ANC and sulfate in lakes requires further analysis within individual watersheds and from a regional scale perspective.

Another significant finding is the small number of low pH, negative ANC lakes with appreciable concentrations of organic anions. Lakes having negative ANC and also associated with the highest organic anion class were found only in the Upper Peninsula of Michigan and the subregion containing Florida and Georgia. The only significant number of high organic anion, low pH lakes sampled occurred in Subregion 3B in the Okefenokee Swamp, Georgia, where lakes were among the most acidic sampled; the minimum pH was 3.81. Overall, in Subregion 3B, 20% of the lakes had $pH \leq 5.4$ and $ANC \leq -7.5$ μeq/L. This demonstrates the potential for organic acids to serve as naturally acidifying sources in some cases. However, a much higher percentage of lakes in the two lowest organic anion classes had negative ANC than in the two highest organic anion classes. In Northeastern Minnesota, which had the highest median organic anion concentration of all subregions 98.6% of the lakes had $pH > 6.0$ and no lakes had $ANC \leq 0$ μeq/L. Although other processes, such as watershed sources of pyrite, can cause a lake to be naturally acidic, the widespread distribution of acidic lakes suggests that a more general mechanism such as atmospheric deposition is likely to be responsible.

The analysis of the relationships between sulfate and ANC, and organic acids and acidic lakes yields similar results from both the U.S. and Canadian surveys. Both countries have regions in which lakes are characterized by high and low acid neutralizing capacity, often coincident with observed patterns of sulfate deposition. However, the lakes in the eastern U.S. generally have considerably higher ANC and pH. This difference can be attributed to the dominant influence of the Precambrian Shield in Canada in contrast with the deeper soils, deeper glacial tills, and smaller aerial extent of igneous bedrock in the eastern U.S. Lakes in the U.S. also exhibit greater regional heterogeneity than those in Canada. For example, bimodal distributions in organic acid classes were observed in Northcentral Wisconsin, Florida, the Upper Peninsula of Michigan and the Upper Great Lakes Area, but not in the Canadian regions.

5. CONCLUSIONS

The ELS-I provides a consistent data base that permits population estimates to be computed for the U.S. regions containing the majority of low ANC lakes. The initial observations are:

a) Acidic and low ANC lakes occur in subregions receiving a considerable range of SO_4 deposition levels. However, the highest percentages of acidic lakes in the Northeast and Upper Midwest are associated with areas receiving the highest sulfate deposition. This association is consistent with the hypothesis that elevated sulfate deposition has caused chemical alterations and acidification in some areas.

b) The large differences in the percentages of low pH and low ANC lakes among subregions in the eastern U.S. receiving similar levels of sulfate deposition indicate that the patterns of atmospheric deposition alone are insufficient to explain fully the distribution of acidic waters without characterizing the underlying sensitivity of the areas.

c) Florida contains by far the highest percentage of acidic lakes in the eastern U.S., followed by the Adirondacks, Southern New England, and the Upper Peninsula of Michigan. The acidic lakes are generally associated with low organic anion concentrations. An adequate explanation is currently unavailable for the high percentage of acidic lakes in Florida.

d) The high percentages of acidic lakes with low concentrations of organic anions is consistent with the observation in Canada that naturally acidic lakes caused by organic acids comprise a small percentage of the total number of acidic lakes. It will be important to identify and define the conditions causing natural acidification.

e) Based on the data presented in this paper, the percentage of low pH and low ANC lakes in the eastern U.S. is far lower than in Canada.

6. ACKNOWLEDGEMENTS

Lockheed Engineering Management Services Co. and Northrop Services, Inc. contributed to the successful completion of the Survey. We would also like to acknowledge the sampling support of EPA Regions 1 (Boston, MA), 2 (Edison, NJ), 3 (Philadelphia, PA), 4 (Atlanta, GA), 5(Chicago, IL), 9 (San Francisco, CA) 10 (Seattle, WA) of the U.S. EPA.

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